Using Formvar to Capture Ice Crystals and Retrieve Roughness Parameters

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Abstract. Ice crystal roughness is a poorly observed and understood parameter, yet it significantly influences crystal's scattering properties and consequently impacts radiative transfer in the atmosphere, contributing to uncertainties in weather and climate forecasting. In this study, we introduce a novel approach as a proof-of-concept to obtain high-resolution roughness measurements, building on the traditional formvar method for capturing ice crystals and validated through comparison of the roughness parameters of salt crystals and salt crystal replicas. Ice crystals were grown in the Manchester Ice Cloud Chamber, collected, and subsequently imaged using various techniques, including a scanning optical profilometer, which enabled the identification of roughness features as small as 0.8 µm. This approach provides critical insights into roughness characteristics that are significant for improving radiative transfer models and forecasts.

1 Introduction

The mean equilibrium surface temperature of Earth is significantly affected by clouds that are responsible for both trapping terrestrial radiation as well as reflecting solar radiation back to space. The balance between these processes determines the change in surface temperature due to changes in the cloud/ presence of the cloud (Hong Y, 2015). As indicated in the IPCC report (2013), the sensitivity in the prediction models for climate changes can reach up to 1.5˚C–2.0˚C. The reason for this range of estimates in the prediction of surface temperature was noted in the report as primarily being due to our limited knowledge of cloud feedback (IPCC, 2013). This lack of understanding of the physical properties of clouds is particularly acute for ice clouds since they are complex over a range of spatial scales and cover large portions of the globe, thus causing significant errors in predictions of climate change. These uncertainties have been substantially mitigated through rigorous, collaborative efforts by the research community, dedicated to advancing the understanding and precise quantification of cloud feedback mechanisms across a range of cloud regimes (IPCC, 2023). As the scientific community has worked to address these uncertainties, our focus here is on issues that can be further mitigated through a deeper understanding of the microscopic properties of ice crystals. This comprises backscattering properties of ice crystals based on their microphysical properties, including shape, orientation, complexity, and roughness.

Cloud models can give accurate results if the inputs represent the actual conditions. Therefore, there have been many efforts over the last 80 years (Bailey, et al., 2009) to understand ice crystal habit formation as a function of environmental conditions, enabling the input to cloud models to be as accurate as possible. In one of the earliest systematic investigations, Magono and Chung (1966) developed a comprehensive classification of natural snow crystal types, creating a foundational framework that has since guided research in snowflake morphology. More recently, in 2005, an experiment was conducted by Libbrecht to explain the mechanism that creates a general hexagonal prism shape for ice crystal growth when their sizes are between 10-100 µm (Libbrecht, 2005). Also, in the same year, Connolly et al. (2005) studied the correlation between the electric field and aggregation of ice crystals and also made observations of chains of ice crystals during fieldwork into deep convective outflows. The results of the study also compared in many aspects with the work done by Saunders and Wahab (1975) since their field data were similar.

As the habit of the ice crystal varies under different conditions, its contribution to scattering behaviour and overall cloud reflection capacity varies. For example, in 2015(a), Smith et al. investigated scattering by hollow columnar crystals. These types of column crystals have hollows in their end faces that alter the scattering behaviour. It was found that the phase functions are not affected too much, but there is a difference in asymmetry parameters as a result of different reflections occurring around the hollows (Smith, et al., 2015a).

Throughout these investigations, the methods used to analyse and observe the ice crystals also developed over time as technology developed. Initially, ice crystals were able to be captured for analysis with a solution often referred to as 'formvar'. Formvar is a solution of polyvinyl formal resin (formvar) dissolved in ethylene dichloride (Schaefer, 1941). The solution is used to coat a surface and then the solvent evaporates leaving behind a plastic coating, or in the case of an ice crystal that melts, a plastic replica. Formvar, often combined with electron microscopy or other imaging techniques, has found widespread application e.g. Kahler et al. (1951) used formvar to coat copper screens for visualizing copper-cystine fibers under electron microscopy. In atmospheric science, following on from Schaeffer (1941), Rucklidge (1965) conducted an experiment that involved the use of the formvar solution to create replicas and examine size and shape properties under an electron microscope. The work of Griggs and Jayaratne (1986) led to improvements in the capture quality, as well as precautions during the application of the procedure. Since then, the method has frequently been used to create and then image replicas, sometimes using high-resolution optical microscopy, as in Smith et al (2015b) who used this technique to classify and explain the habit of crystals grown in different conditions. Finnegan and Pitter (1988) investigated the replica process to explain aggregation and secondary ice formation. In the study of Miloshevich et al. (1996), formvar was used to collect cloud particles as a part of the process for a balloon-borne cloud particle replicator to measure the vertical profile of particles. Another replicator was designed and used by Warburton et al. (1983) for the study of transmission of snowfall. This replicator also included a formvar to capture crystals by creating a thin film and allowing crystals to impact it to produce detailed imprints on the film. More

recently, the use of formvar for crystal habit classification has been largely superseded by optical imaging probes that offer automatic sizing and shape determination but at a lower resolution than that available from standard optical imaging of formvar replicas. These probes provide fast and in-situ imaging, but the resolution of the images is not adequate to analyse the surface details or roughness.

Developments of the original optical imaging probes such as the cloud particle imager (CPI) have allowed roughness to be inferred from the light scattering patterns off cloud particles using instruments such as the small ice detector (SID-3; Ulanowski et al. 2014). This technique measures the combined effect of small-scale surface roughness and the crystal complexity as the overall practical impact of 'roughness' on scattering but does not directly measure the small-scale roughness features. This method, and variations on SID-3, have been used airborne to measure within cirrus (Ulanowski et al. 2014; Järvinen et al. 2018), and in the laboratory to determine the impact of growth conditions on crystal roughness (Voigtländer et al. 2018). Meanwhile Magee et al. (2014) grew ice crystals in the low-pressure chamber of an Environmental Scanning Electron Microscope (ESEM) and observed a wide range of surface features on scales from 100 nm to more than 10 µm on both these and externally grown crystals transferred to the chamber. Meanwhile Collier et al. (2016) used a sand particle scanned with an Atomic Force Microscope (AFM) as a proxy for ice particle roughness parameters. The roughness values obtained from the sand grains were found to be similar to those retrieved by SID-3 for ice crystals, making sand a suitable medium for capturing roughness then applied to scattering models.

Up to this point, a direct measurement of the surface roughness of ice crystals has been elusive, yet the traditional formvar technique has the potential to enable such measurements. There is a pioneering study that investigates the formvar replica process and its ability to capture roughness details (Agar, et al., 1956). This study used formvar solution to coat metal alloys, and an interferometer microscope was used to conduct further roughness measurements. Ideally, a uniform thickness of formvar should be present across the surface. On average, the thickness of the formvar solution achieved was around 700 Å. After the coating process, scans of the reverse of the replica and the original material were compared, and the difference between them was found to be negligible.

To move beyond having to use proxies of roughness, in this study, we demonstrate a technique to obtain a direct measurement of the roughness of ice crystals by coupling the relatively old method of capturing ice crystals in formvar with newer instrumentation in the form of an optical profilometer. Nevertheless, using such an old technique requires some modifications to improve the quality of capture and allow for detailed imaging that then enables a direct measurement of the roughness and provides a useful complementary technique to the current suite of cloud imaging probes. Unlike the method described in Smith et al. (2015a), we introduced modifications such as increasing the amount of formvar solution used, which helped sustain a more consistent coating. Additionally, the process was carried out under a vacuum to make sure that the replicas were able to be created whose details will be explained in the methodology section.

2 Methodology

The formvar replica technique has traditionally been an efficient way of capturing ice crystals and preserving their surface structure, which is particularly important for ice crystals which would otherwise melt once collected and brought to laboratory temperature. While it has successfully been used for determining crystal habits and features such as hollowness, identifying roughness of a crystal requires evidence that the formvar can accurately reproduce the necessary high-resolution detail. We present the final methodology developed to determine the scale of roughness of ice crystals. The results section will also illustrate some of the intervening stages in developing the technique, for example the imaging of salt crystals as proof of concept that the formvar replicas could maintain structure of the magnitude expected of ice crystal roughness.

The initial step of the traditional method is the placement of a solution of 0.6 wt% polyvinyl formal resin in chloroform (CHCl3) by using a brush onto a glass slide that is then used to capture the ice crystals. Once the formvar solution has subsequently dried out, a replica of the ice crystal is left in the solid formvar which can be investigated under microscopy to retrieve features of the crystal.

The brush leaves a thin layer of solution after it is applied to the slide. This has the advantage of drying rapidly but the crystals cannot be covered fully. This results in imprints of crystals which are suitable for habit identification but not ideal for roughness detection. A more effective approach involves capturing the entire crystal by suspending and encasing it in formvar, thereby ensuring that all surface cavities remain filled during the solidification of the solution and the sublimation or ablation of the crystal. As most of the crystals generated in the Manchester Ice Cloud Chamber (MICC) fall under 1 mm in size, that depth of solution was targeted for the experiment, but brings its own challenges.

In order to achieve the increased thickness of formvar solution required, aluminium foil was used to create a wall around the glass slide, creating a pool of the solution to a desired level of thickness. It is critical to have enough depth for ice crystals to submerge fully but the evaporation of the chloroform solvent from a thicker layer of formvar, at the low temperatures required for crystal formation, proved too slow at standard atmospheric pressure. Early trials found that the chloroform could not completely evaporate before the crystals deformed or ablated.

The study of Kawamura et al (1987) shows the evaporation rate per unit area is calculated following Eq. (1):

$$E = k * M * P(T_s)/RT \quad (1)$$

Where k is the mass transfer coefficient, M is the molecular weight, $P(T_s)$ is the vapour pressure at T_s which is the temperature of solution surface, while R and T are the gas constant and absolute temperature of air respectively. The evaporation rate is directly proportional to vapour pressure, thus at a given temperature increasing the vapour pressure increases the evaporation

rate. As seen in Figure 1 at a temperature of –20˚C, the vapour pressure of chloroform is decreased by a factor of at least 5.5 when compared to room temperature.

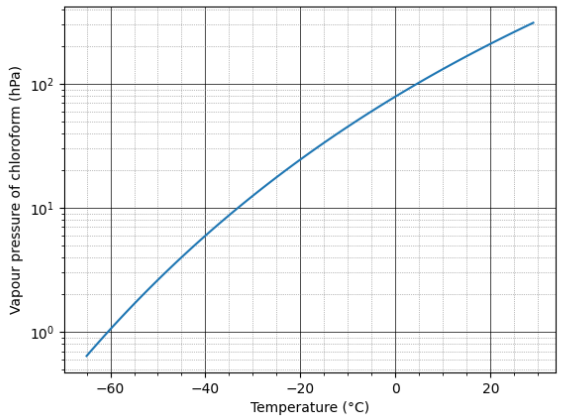

Figure 1. Vapour pressure of chloroform (hPa) vs temperature (˚C) adapted from Cheric (1995)**.**

This suggests a solution to the problem caused by slow evaporation of chloroform: that by reducing the external pressure on the slide the evaporation rate would be increased. Accordingly, the formvar slide, with its captured ice crystals was sealed inside a bell jar and air slowly pumped from within. The target pressure was 140–200 hPa achieved at a flowrate (set to 10 m^3min^{-1}) that would not disturb the sample contained within the bell jar.

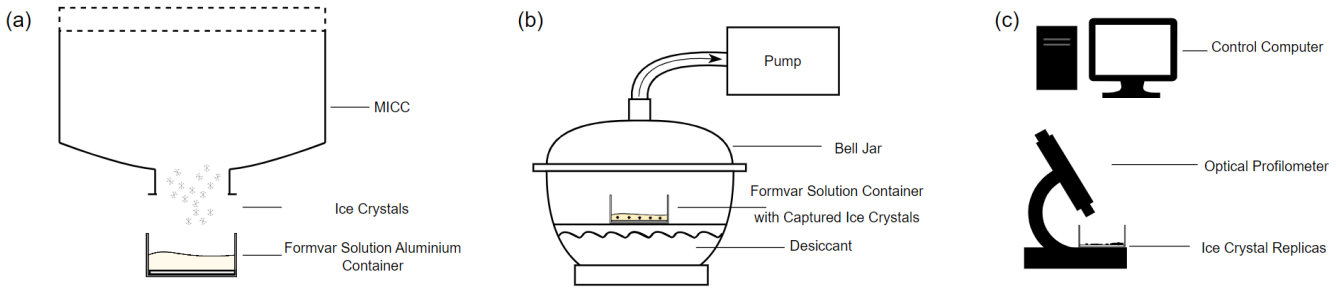

Figure 2. Schematic (not to scale) showing steps described in methodology: **(a)** sample collection, **(b)** evaporation process, and **(c)** optical profilometer scanning.

At this flowrate, it took a few minutes to reach the desired pressure level, and once it was reached, the evaporation visually inspected: as the evaporation occurs, the colour of the solution goes from transparent to silky white. The sample was left to evaporate for 10 minutes to ensure the process was complete.

Samples were initially examined using an optical profilometer, specifically the Keyence X200K 3D Laser Microscope with a lateral resolution less than 10 nm and vertical resolution less than 5nm, to ensure that the crystal replicas remained intact without undergoing any additional deformations. This preliminary inspection enabled the identification of areas of interest for more detailed analysis. Once selected, these areas were thoroughly surface scanned using the same instrument, and the resulting scan data was stored for further evaluation. The primary advantage of utilizing the optical profilometer over earlier imaging techniques lies in its dual functionality. It offers a rapid initial optical assessment, followed by a highly detailed 3D surface scan that captures intricate topographical features with precision. The optical profilometer, such as a laser scanning confocal microscope, utilizes non-contact techniques to measure surface profiles, roughness, and thickness across different materials. It employs both laser and white light sources to simultaneously capture laser intensity, colour, and height data, allowing for the generation of fully focused colour images and detailed 3D height profiles. In contrast to traditional methods like scanning electron microscopy, which may distort samples due to electron beam interaction, the profilometer maintains sample integrity while delivering high-resolution data. This capability makes it well-suited for both quick assessments and comprehensive surface analysis, providing excellent versatility and accuracy, compared to other methods used in the early stages of technique development. After the scanning data was retrieved, it was examined and analysed using a software package called Gwyddion. This program allows users to import surface scanning data and to focus on extracting detailed data from a specified area of the scanned surface, in this case the crystal replicas and not the surrounding formvar, though background areas of formvar were scanned to provide a baseline roughness value. It enables parameters like mean roughness to be calculated following Eq. (2):

$$R_a = \frac{1}{L}\int_0^L |Z(x)|dx \quad (2)$$

Where L is evaluation length and Z(x) is height profile (Gadelmawla, et al., 2002) and root mean square (RMS) roughness is calculated following Eq. (3):

$$R_q = \sqrt{\frac{1}{L}\int_0^L Z(x)^2 dx} \quad (3)$$

3. Results and Discussion

The methodology outlined in the previous section was developed to demonstrate that it is possible to extract roughness information directly from ice crystals formed within clouds. It was inspired by the observation that traditional optical microscope images are not sufficiently detailed to provide information on roughness as the required high image resolution cannot be achieved. During the initial examination of the optical microscope sample images retrieved using the brush technique, there were features observed that could be artefacts of the formvar process itself, rather than features of the original crystals. Clarifying whether these are artefacts or belong to the crystals themselves, however, requires high-resolution images.

Initially, scanning electron microscopy (SEM) was employed to capture the high-resolution images necessary for the analysis by scanning the surface of the material with a focused beam of electrons. SEM was chosen for its ability to achieve resolutions of up to approximately 100 nm, which theoretically provides sufficient detail to meet the 100nm roughness measurement goal. While SEM was not used directly for surface roughness measurement purposes in this study, it played a critical role in the preliminary phase by allowing for detailed characterization and identification of artefacts introduced during the preliminary formvar replication. The ability to distinguish artefacts from actual microstructural features was important for designing the final experimental method and ensuring more accurate measurements in following stages of the research.

In order to explore this further, small areas of interest were chosen from the optical microscope examination and prepared for SEM imaging. Once each area is chosen, the whole slide was covered with foil, leaving a small hole approximately ~1mm^2 in size in the foil which defines the area of interest (Figure 3). Since scanning might need to be repeated several times to get images with different resolutions and to prevent causing damage to the whole slide, only a small portion of a slide was prepared for imaging by coating with gold, and the scanning then carried out to image the selected feature.

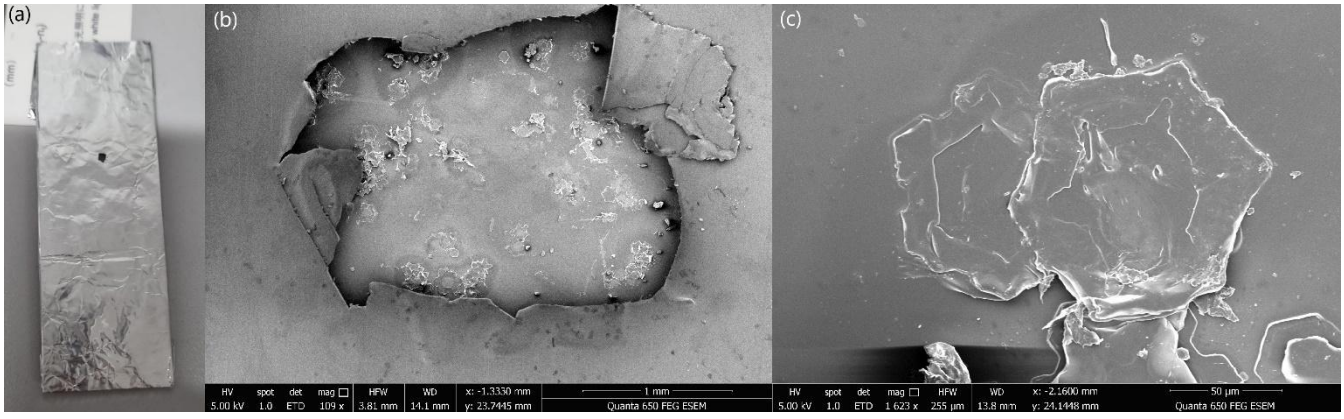

Figure 3. SEM imaging steps described: **(a)** sample 73 covered with foil with an opening on it, **(b)** the selected area's SEM image and **(c)** close-up image of 2 hexagonal plates that sit on each other.

In total, eight samples were prepared for this preliminary investigation, and were imaged with varying zoom scales during three different SEM sessions. When those images were examined thoroughly, it can be seen that a significant portion of the features are artefacts of the formvar process (Figure 4), that is the crystals did not form complete 3-dimensional replicas. One reason for this was considered to be the thin layer of formvar delivered by brushing it onto the slide, so there was insufficient depth for the crystals to be fully covered, or the shape and density of crystal could have prevented it sinking sufficiently for full cover. Finally, we had to consider whether the formvar material might start to deform after or whilst the ice crystals evaporated. A relatively simple workaround for much of these problems was to increase the depth of the formvar solution layer

by constructing a container to enable a pool of formvar to cover the slide, deep enough to capture the majority of single crystals, though it has consequences in terms of the evaporation rate as discussed previously.

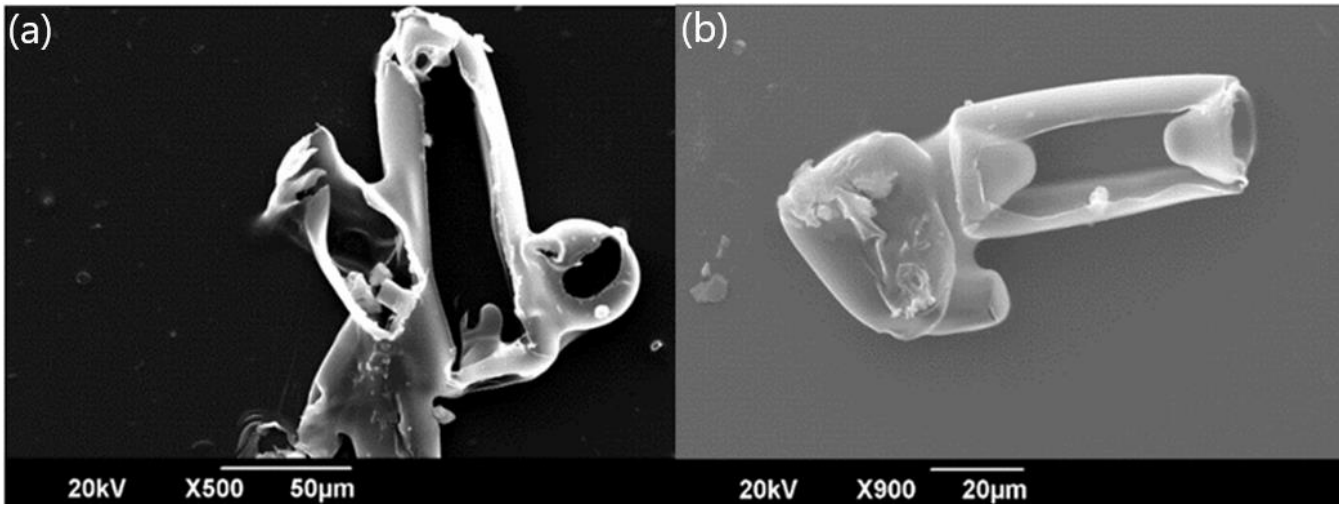

Figure 4. SEM images of the ice crystals which did not sink fully into the formvar solution and consequently each replicas exhibits a hole in its upper surface.

3.1 Validation using salt crystals

Though the procedure described in Section 2 allowed us to measure the roughness of atmospheric ice crystals, an essential step is to ensure that the roughness values obtained from a replica are the same as would be measured from the crystal itself, that is to confirm that the formvar replica did not deform. To this end, the process was repeated with table salt (sodium chloride) crystals as a validation source, though with slight adaptations. Initially, the brush technique was employed to create replicas to investigate surface structure. However, after analysis with optical profilometer, we found that the remaining surface details were incomplete or corrupted as shown in Figure 5 and did not provide any useful information about the surface characteristics, leading us to refine our approach. Therefore, we applied the formvar pool method discussed in methodology to produce further samples.

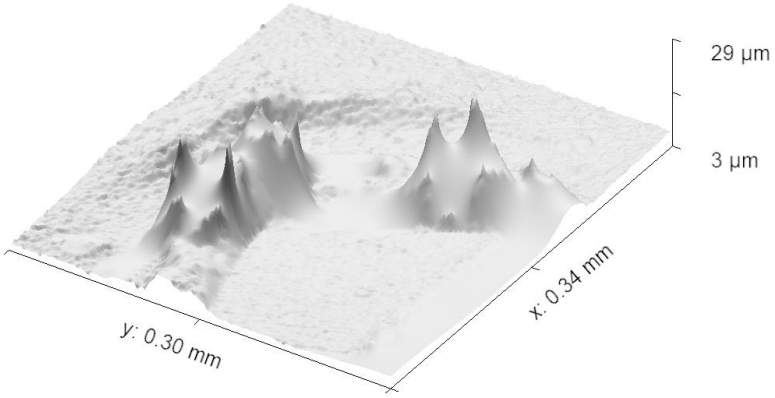

Figure 5. Surface scanning, using an optical profilometer, of a replica of salt crystal created by using brush technique. The image reveals incomplete or distorted surface details due to limitations of the brush technique, which failed to capture fine structural features accurately. This led to the refinement of the replication method

Once the chloroform had evaporated from the formvar solution, the formvar leaves behind a replica of the salt crystals. At this stage though, the salt crystals remain and so must be removed by being dissolved in water. For this the sample slide was gently placed in a water bath and left for 15 minutes until the salt dissolved and could be washed away. With the salt removed from the sample, only the replica shell remained, and it was scanned with the SEM microscope. In addition, salt crystals from the same batch were placed directly on a microscope slide and imaged and analysed in the same manner.

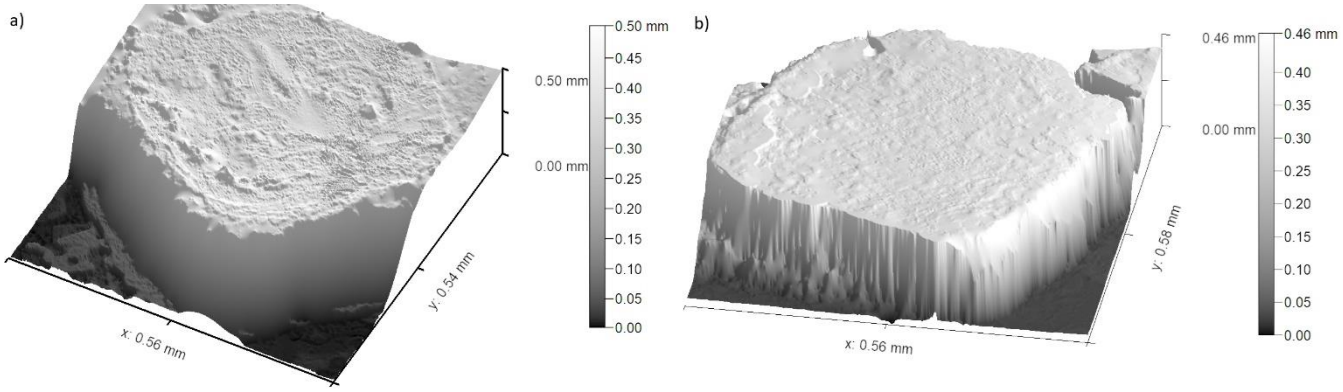

Figure 6. (a) Scan of a randomly chosen salt crystal without formvar solution application. **(b)** Scan of a randomly chosen formvar shell after salt crystal removal. Both of them were taken with an optical profilometer.

The image in Figure 6 (left), shows the result scanning of a sample salt crystal without the application of formvar whereas the right-hand side of the figure shows the result of the replica formvar process. In a comparison of both images, it can be seen that a sufficient depth of formvar allows the full surface of the crystal to be imaged and complete scan of the surface retrieved.

This demonstrates our method's ability to capture the surface features effectively and provides a clear comparison of replicated surfaces despite the fact the sample salt crystals were not those subsequently covered in formvar and were randomly chosen. The results of the comparative roughness analysis of four salt crystals and four independent replicas are shown in Table 1.

From these, we can see that the formvar replica process retains the roughness parameter measurement of the original crystal with a good level of accuracy. The deviation between the two samples is less than 5% which gives confidence that the measurement technique can represent true roughness parameters when it is applied to ice crystals. It should also be noted that salt crystals exhibit a range of roughness values, with the largest being approximately twice as large as the smallest in our samples, and this range is also seen in the replicas. This variability in roughness is also reflected in the replicas, despite the random selection of salt particles for measurement. These results provide confidence that the formvar replica technique can be applied effectively to characterize roughness parameters of ice crystals.

Table 1. 1D Roughness parameters of four salt crystals and four salt crystal replica samples.

	Salt Crystal	Replica of Salt Crystal
Area	0.104 mm^2 to 0.255 mm^2	0.1124 mm to 0.218 mm^2
RMS Roughness	13.27 µm to 26.74µm	10.63 µm to 22.73µm
Mean Roughness	10.76 µm to 19.16 µm	8.27 µm to 18.96 µm
Skewness	–1.447 to –0.35	0.304 to 0.454

The linear profiles used for roughness measurement were selected by making the scans for the full surface of each crystal, to make sure that all surface features were included. Although increasing the sample size would provide more robust statistical confidence, the use of four salt crystals provides a reasonable basis for comparison as they were covering full surface features. Skewness was measured using Gwyddion software, which calculated the surface asymmetry based on the height distribution of the features across the scanned profile, providing additional insight into the surface texture. A positive skewness indicates a surface dominated by peaks, while a negative skewness suggests deeper valleys. The observed skewness differences highlight variations in surface texture, which may influence properties such as adhesion, wettability, or mechanical interactions. Also, as a final check to make sure that these values are not significantly affected by the presence of formvar itself, a formvar surface with no crystals present was also scanned and analysed, the RMS roughness in this case being found to be 175 nm. This is orders of magnitude less than the values obtained in Table 1, supporting our assertion that the parameters retrieved from replicas are close to those for the original crystals.

3.2 Retrieval of Ice Crystal Roughness Parameters

At this stage, roughness parameters for ice crystals produced in the Manchester Ice Cloud Chamber (MICC) were explored following the procedures described in the methodology section. As a proof of concept for an experimental technique, rather

than a full treatise of ice crystal roughness, a single temperature (–20°C) was chosen for cloud formation and an ice cloud was produced. In the cloud generation chamber, temperature and humidity are carefully controlled to initiate condensation under a supersaturated environment, allowing water vapor to condense and form ice crystals and throughout this study, only one set of environmental conditions is used. These crystals then fall and exit through an opening at the bottom of the chamber, where they were captured, and replicas were created. The replicas were inspected with an optical profilometer rather than the SEM described for early investigations due to improved speed, access and non-contact aspects of the method.

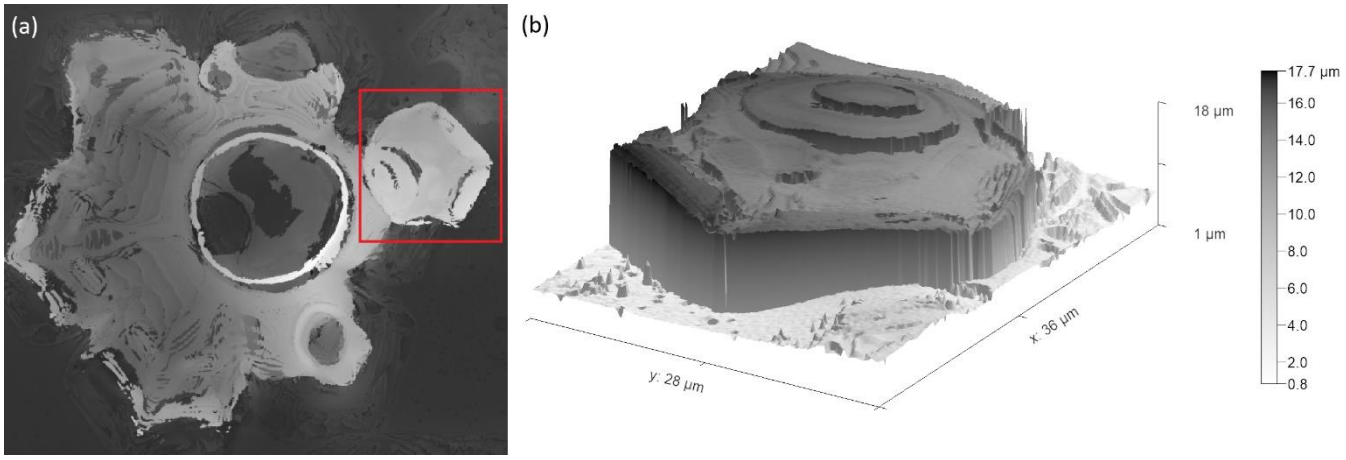

Figure 7. (a) One plate sits on another crystal marked with a red square. **(b)** Detail of surface scan of crystal highlighted by red square. Both scans were taken with an optical profilometer.

An example crystal replica is shown in Figure 7, visually demonstrating that it is possible to capture the detailed surface features and shape of ice crystals with formvar. Multiple ice crystals were analysed using the same software, Gwyddion, to calculate roughness parameters and ascertain a range for the different variables. These results are shown in Table 2.

Table 2. Roughness parameters of three ice crystal replicas.

	Ice Crystal Replica
Area	300 µm^2 to 800 µm^2
RMS Roughness	1.2 µm to 3.6 µm
Mean Roughness	0.8 µm to 2.66 µm
Skewness	-2.36 to 0.28

In using formvar to create ice crystal replicas, we were aware of the process of chemical etching which has been recognized as a potential source of error in formvar replicas, as discussed in the previous studies (Sinha, 1977, 1978). This occurs when water molecules which are weakly bonded can go into the formvar solution and leave artefacts called “etch-pits” that could

then be erroneously identified as crystal roughness . While we acknowledge these artefacts as a critical potential factor, we did not observe etching pits in our analysis.

Roughness parameters were then compared over 35 randomly chosen areas from four different crystal surface scans to establish a correlation with the length parameter (side of the area scanned). By selecting various regions of the ice crystal surface, roughness can be analysed at different scales to provide a more detailed assessment of surface variability. In this study, multiple areas of different length parameter were chosen to capture localized variations, and their roughness values were statistically compared in order to consider different types of structural feature. The results, presented as a box plot in Figure 8, demonstrate the roughness distributions for each selected area, highlighting trends and potential correlations with the length parameter.

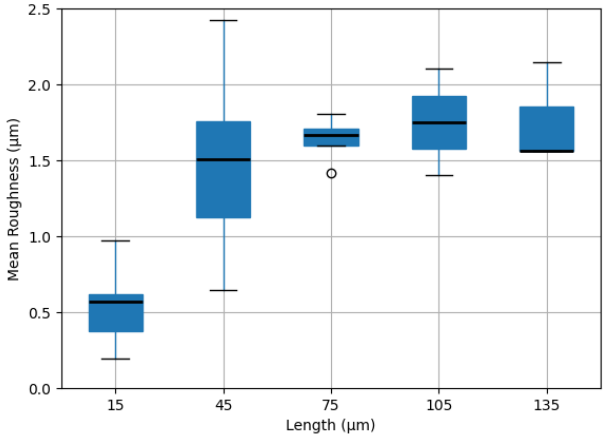

Figure 8. Statistical comparison of length (µm) vs. mean-roughness values (µm) across multiple selected areas of multiple ice crystals' surfaces (n=7 for each size bin).

In general, this distribution can be further simplified to show the correlation between length and roughness parameters. As the length increases, less frequent and larger features on the ice crystal surface are more likely to be captured and correspond to higher roughness values. The mean roughness plateaus at about 1.7 um, indicating a limit to the small-scale roughness, with larger surface areas not capturing any greater roughness features.

As before with the salt crystal analysis a range of roughness values are seen in the samples, though the range among means-roughness of size bins is not quite so large for our ice samples, only varying by a factor of 1.5–1.8 for over most of the size bins. However, we acknowledge that the environmental conditions are expected to have a significant effect on the roughness of ice crystals, particularly the supersaturation of the environment in which they form, and we have presented results from a single set of controlled conditions in developing this technique. Collier et al. (2016) used the roughness parameters of sand particles as a proxy for model work for asymmetry parameter calculations, yet it is encouraging that findings are relatively

close to the parameters in that work. Their two-scale roughness approach was applied to crystals with a size of 32.5 µm, which lies between our lengths 15 and 45 µm. The mean roughness for the large-scale features was measured at 1.08 µm, while the mean roughness for the small-scale features was measured at 0.13 µm which is approximately the lowest value that we observed with our smallest length scale of 15 µm. In any event roughness will significantly affect the scattering behaviour of the crystals (Collier, et al., 2016), and this is often an underappreciated effect. In simple terms it can explain the relative rare of observance of halos in thin cirrus (Smedley, 2003; Forster and Mayer, 2022).

We stress that our study is a proof-of-concept study, and we anticipate that ice crystal roughness will vary with temperature, habit, and supersaturation. This variability is especially significant in high-altitude cirrus clouds and turbulent cumulus clouds, where the irregularities in ice crystals or droplets can lead to increased roughness, significantly impacting light scattering and the overall optical properties of the clouds. The high roughness can lead to higher scattering efficiencies and smaller asymmetry parameters (Ulanowski, et al., 2006), affecting the amount of solar radiation reflected back into space and complicating satellite retrievals of cloud properties. The method presented here can be repeated under varying conditions which can provide an effective way to map the surface characteristics of ice crystals across different environmental conditions. By applying this technique to varying atmospheric settings, we can better understand how roughness evolves, which is essential for improving satellite observations, enhancing cloud property retrievals, and refining climate models that rely on precise knowledge of cloud dynamics.

While this method was developed in a laboratory setting, it could potentially be adapted for airborne campaigns, once challenges of delivering the cloud particles to a protected formvar slide are overcome. The lower pressure available at altitude will assist rapid enough drying of the formvar, could be aided by slightly thinner coatings. Additionally, this method can be employed at some high-altitude ground-based stations situated at cloud level, allowing for the collection of samples directly from natural clouds. Such efforts are essential, as they can improve our understanding of cloud feedback mechanisms and the intricate details of roughness, ultimately contributing to a more comprehensive understanding of the net energy balance in the atmosphere.

This method offers high-resolution and provides detailed structural information on ice crystal surface roughness despite the manual input required and the relatively limited ice crystal samples, compared with optical instruments like SID-3. While SID-3 is effective for airborne measurements, it gives an indirect measure of structure and roughness, while our offline approach allows for more precise analysis of surface roughness in a controlled environment. The two methods complement each other, with laboratory work providing detailed insight into ice crystal surface structure that can enhance future airborne studies.

Lastly, in terms of general roughness, providing similar results between actual crystal and its replica is important to ensure that this method can be used as a reliable proof-of-concept for future studies. While our current analysis simply uses one-

dimensional roughness data, this study demonstrates the potential of this method to be improved and expanded to capture more comprehensive roughness characteristics. The consistency in roughness trends observed, despite the 1D measurement limitations, suggests that this method can be the basis for understanding ice crystal surface roughness. By building upon these results, we anticipate that future modifications in this technique will enable more precise evaluations by extracting complex roughness information to express crystal surface complexity under varying conditions.

4 Conclusion

According to the IPCC report (2013), cloud models are inadequate for informing climate change models, which hinders the ability of models to provide accurate climate change predictions. Improvements are necessary in many aspects, especially with regard to their microphysical properties. The microphysical properties directly impact scattering characteristics. By representing the conditions of ice clouds accurately within models, reliability in predictions can be enhanced. While previous studies have extensively examined the shape and habits of ice particles, this study solely concentrates on a method for roughness parameter retrieval, leaving exploration of the effect of environmental conditions on roughness formation to future investigations.

In calculating roughness parameters, this formvar technique, combined with modern imaging opportunities, offers novel possibilities for direct measurement of the roughness, despite its being based on an older technique. Roughness makes a critical contribution to scattering properties. Therefore, accurately determining roughness parameters can improve the prediction of scattering outcomes, which in turn can enhance cloud models and make their feedback to climate models more precise. Our method has resulted in the successful creation of ice crystal replicas, and their imaging and analysis at roughness scales by profilometer. The accuracy of the roughness parameters derived was validated through a comparative analysis using salt crystals, where the integrity of the crystals could be maintained independently of the formvar. This indicated that the roughness parameters found with our methodology have an uncertainty of no more than 5-6%, where some of this uncertainty may be natural variation in the salt crystals.

A critical aspect of this study lies in its innovative integration of established techniques with cutting-edge instruments, which enables the extraction of valuable information on ice crystal roughness parameters. This research provides a detailed method for capturing and visualizing ice crystals captured in formvar using an optical profilometer, ensuring that key surface characteristics are preserved. Through comprehensive scanning, we have measured the roughness parameters. Although the current work does not directly show the applicability of these one-dimensional roughness values to two-dimensional anisotropic rough surfaces or optical property calculations, it can be a foundation for future studies to provide such roughness parameters to be used in models and enhancing our understanding of cloud microphysics and their role in climate analysis. These findings can be also compared with both laboratory data and in-situ measurements, offering new perspectives on cloud microphysics. This work not only enhances our understanding of cloud behaviour but also contributes to refining models that simulate cloud properties, which are crucial for weather predictions and climate analysis. By bridging the gap between theoretical models and real-world observations, this study plays a key role in advancing our comprehension of cloud dynamics and their influence on the Earth's climate system.

Code Availability

The Python code used to in this study is available from the corresponding author upon request.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Author Contribution

Omer Celebi conceptualized and designed the study, conducted all data collection, analysis, and interpretation, and drafted the manuscript. Ann R. Webb, Paul Connolly and Andrew R.D. Smedley provided guidance, supervision, and critical revisions of the manuscript.

Competing interests

The contact author has declared that none of the authors has any competing interests.

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